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PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Yoo et al.
Serial No. : 09/581,804 Examiner : R. A. Lee
Filed : June 16, 2000 Art Unit : 1713
For : THERMOPLASTIC TRANSPARENT RESIN COMPOSITION AND
METHOD OF MANUFACTURING SAME

**SUBMISSION OF ORIGINAL DECLARATION
UNDER 37 C.F.R. SECTION 1.132**

I hereby certify that this paper is being deposited with the United States Postal Service as first class mail in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

October 18, 2003

Date of Deposit

Bradley B. Geist

Attorney Name

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27,551

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Date of Signature

Commissioner for Patents
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Sir:

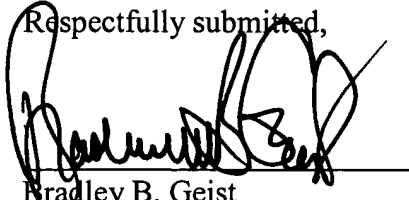
This paper and the attached Declaration is submitted in connection with, and in support of the amendment and Request for Continued Examination filed on May 14, 2004.

Prosecution was requested to be suspended, and this request was granted.

PATENT

The Commissioner is authorized to charge any fees associated with this communication to Deposit Account No. 02-4377.

Respectfully submitted,

A handwritten signature in black ink, appearing to read 'Bradley B. Geist', is written over a horizontal line.

Bradley B. Geist
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Enclosure



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of : Yoo *et al.*
Serial No. : 09/581,804 Art Unit : 1713
Filed : June 16, 2000 Examiner : Lee, R.
For : THERMOPLASTIC TRANSPARENT RESIN
COMPOSITION AND METHOD OF
MANUFACTURING SAME

DECLARATION UNDER 37 C.F.R. § 1.132

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Assistant Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

I, Chan-Hong Lee, do declare:

1. I am a co-inventor of the invention disclosed in the above-identified application.

2. I am a citizen of Korea and I reside at LGHana Apt 102-901, Shinseong-dong, Yuseong-gu, Daejeon, Korea In 1978, I graduated from the University of Seoul in Seoul, Korea with a Bachelor's Degree in Chemical Engineering. In 1988, I graduated from the University of

Oklahoma in Norman, Oklahoma, USA with a Doctorate Degree in Chemical Engineering.

Since 1988, I have been working in the field of Emulsion Polymerization. I have been employed by LG Chemical Ltd. since 1988, and part of this time has been spent in the field of polymer chemistry. Since 1997, I have been developing Transparent ABS Resin in the ABS Resin Development Department of LG Chemical Ltd. I have been granted 23 patents on emulsion polymerization and I am the author of four papers published in academic journals in the field of emulsion polymerization. I am familiar with pending United States Patent Application Serial No. 09/581,804 and with the following references cited by the Examiner during the prosecution of this same application:

- a. U.S. Patent No. 4,767,833;
- b. JP 56-41216;
- c. U.S. Patent No. 5,200,441;
- d. U.S. Patent No. 5,225,494;
- e. U.S. Patent No. 5,071,946;
- f. U.S. Patent No. 3,880,786;
- g. U.S. Patent No. 3,855,355;
- h. U.S. Patent No. 6,080,815;
- i. U.S. Patent No. 4,703,090;
- j. U.S. Patent No. 4,581,408;
- k. U.S. Patent No. 4,520,165; and
- l. International Patent Publication WO00/26298

3. The invention disclosed in the above identified application relates to compositions

of transparent thermoplastic resins and methods for the manufacture of the same. Specifically, the invention relates to compositions and methods of manufacture for a transparent thermoplastic resin composition comprising 3-15 parts by weight of small aperture polybutadiene rubber latex; 5-25 parts by weight of large aperture polybutadiene rubber latex; 40-70 parts by weight of a methacrylic acid alkylester compound or an acrylic acid alkylester compound; 15-30 parts by weight of an aromatic vinyl compound; and 1-20 parts by weight of a vinylcian compound, wherein the small aperture polybutadiene rubber latex has a particle diameter of 600-1500 Å, a gel content of 70-95%, and a swelling index of 12-30, and the large aperture polybutadiene rubber latex has a particle diameter of 2600-5000 Å, a gel content of *greater than 80 and not exceeding 95%*, and a swelling index of 12-30, and wherein the total refraction coefficient of the composition, excluding the polybutadiene rubber latex is between 1.510 and 1.526 (emphasis added).

4. As presently claimed in pending U.S. Patent Application Serial No. 09/581,804, the gel content of the large aperture rubber lies outside of the range of gel content for large aperture rubber latex taught by Yumoto *et al.*, who note that “component (B) [(large aperture styrene-butadiene rubber)] *is required to have a gel content of 10 to 80% by weight*” (emphasis added). When the gel content of large aperture rubber latex is below 80%, the swelling index is higher and thus monomers grafted in the rubber swell more. Furthermore, the extent to which the grafted monomers swell is variable. As a result, it is difficult to achieve high transparency in thermoplastic resins produced from large aperture rubbers with low gel contents.

The relationship between gel content of the large aperture rubber and transparency of the resin is directly demonstrated by the following experimental evidence. Three types of large

aperture polybutadiene latexes, each having a different gel content, were produced by the processes set forth below. All three large aperture rubber latexes possessed a particle size of 3,000 Å.

A. Polybutadiene 1

Small aperture rubber latex was produced according to the process of Example 1 of U.S. Patent Application Serial No. 09/581,804, except that the reaction was performed at 76°C rather than 65°C and tertiary dodecyl mercapta (TDDM), which is a molecular weight-controlling agent, was added in an amount of 0.2 parts by weight rather than 0.3 parts by weight. Large aperture rubber latex then was produced from this small aperture rubber latex as described in Example 1 of U.S. Patent Application Serial No. 09/581,804. The gel content of the large aperture rubber latex was 95%.

B. Polybutadiene 2

Large aperture rubber latex was produced according to the process of Example 1. The gel content of the large aperture rubber latex was 85%.

C. Polybutadiene 3

Small aperture rubber latex was produced according to the process of Example 1 of U.S. Patent Application Serial No. 09/581,804, except that the reaction was performed at 61°C rather than 65°C and tertiary dodecyl mercapta (TDDM), which is a molecular weight-controlling agent, was added in an amount of 0.4 parts by weight rather than 0.3 parts by weight. Large aperture rubber latex then was produced from this small aperture rubber latex as described in Example 1 of U.S. Patent Application Serial No. 09/581,804. The gel content of the large aperture rubber latex was 70%.

Thermoplastic resins were produced by performing graft copolymerization based on the same materials and processes of Example 1 of U.S. Patent Application Serial No. 09/581,804. Table 1 presents the relationship between the gel content of the large aperture polybutadiene used to produce the resins and the haze values of the resins produced from the various types of large aperture polybutadiene.

Table 1.

	Gel Content (%)	Haze Value (%)
Polybutadiene 1	95	2.0
Polybutadiene 2	85	2.0
Polybutadiene 3	70	5.9

When large aperture polybutadiene has a gel content of less than 80%, which is outside of the range claimed in U.S. Patent Application Serial No. 09/581,804, the haze value of the resin is high. Such a resin has poor transparency and therefore is unsuitable for use as a transparent thermoplastic resin.

The only instance in Yumoto *et al.* disclosing large aperture rubber with a gel content greater than 80% is Comparative Example 14, which appears in Table 1 of Yumoto *et al.* As noted previously, Comparative Example 14, in which the large aperture rubber has a gel content of 90%, also possesses a Haze value of 10%, which is unacceptably high for a transparent thermoplastic resin. In contrast, the transparent thermoplastic resins of the instant invention, with gel contents of greater than 80 and not exceeding 95%, have Haze values of 1.5-4.5%.

On the basis of the explicit statement by Yumoto *et al.* that their large aperture rubber is required to have a gel content of no more than 80%, and the fact that the Comparative Example

in which the large aperture rubber has properties that are wholly inconsistent with a transparent thermoplastic resin of the instant invention when the gel content is greater than 80%, I declare that one of ordinary skill in the art of transparent thermoplastic resins would not find it desirable to employ large aperture rubbers with gel contents of greater than 80% for the production of transparent thermoplastic resins. Thus, the person of ordinary skill in this field, considering the teachings of Yumoto *et al.* in their entirety, would conclude that large aperture rubber with a gel content of greater than 80% would be unsuitable for use in transparent thermoplastic resins. This person, therefore, would have no motivation to combine the teachings of Yumoto *et al.* with those of JP 56-41216 to replace the styrene-butadiene rubber of Yumoto *et al.* with the polybutadiene of JP 56-41216. Even should one do so, the resulting product does not teach or suggest all of the limitations of the claims of the instant application. Consequently, I declare that the invention of the instant application is not rendered obvious by the combined teachings of Yumoto *et al.* and JP 56-41216.

5. The Examiner asserts that the teachings of Yumoto *et al.* in combination with JP 56-41216 still render the compositions and methods of the instant invention obvious, despite the importation into the claims of a limitation with regard to the swelling index, which is not taught or suggested by either reference. According to the Examiner

[t]he combination of reference[s] does teach the chemical constituents of the claimed composition, as well as the relative amounts. The reference is simply silent with respect to physical properties. Since chemical compositions and the associated physical properties are not mutually exclusive, compositions which are essentially the same are expected to exhibit essentially the same properties. In this respect, the burden of proof was shifted to Applicants to establish unobvious differences in the claimed physical properties, but to date, this duty has not been fulfilled

I first note that, as presently amended, the compositions claimed in the instant application are no longer "essentially the same" as the compositions taught or suggested by Yumoto *et al.* alone or in combination with JP 56-41216. Thus, one of ordinary skill in the art would not have an expectation that the compositions would possess the same physical properties with regard to the swelling index.

In general, as gel content becomes lower, the swelling index becomes higher. Since the gel content of the large aperture rubber in Yumoto *et al.* is 10-80%, a swelling index of the rubber would be between 30-100. When rubber has a high swelling index, monomers grafted in the rubber swell more, and the extent to which the grafted monomers swell will be variable. As a result, regulating the refraction coefficient of the rubber and the monomers grafted therein will be difficult. Accordingly, the transparency of the resins of Yumoto *et al.* is limited due to the high swelling index.

The relationship between the swelling index and transparency is directly demonstrated by the following experimental evidence. Three types of large aperture polybutadiene latexes, each having a different swelling index, were produced by the processes set forth below. All three large aperture rubber latexes possessed a particle size of 3,000 Å.

A. Polybutadiene 1

Small aperture rubber latex was produced according to the process of Example 1 of U.S. Patent Application Serial No. 09/581,804, except that the reaction was performed at 75°C rather than 65°C and tertiary dodecyl mercapta (TDDM), which is a molecular weight-controlling agent, was added in an amount of 0.2 parts by weight rather than 0.3 parts by weight. Large aperture rubber latex then was produced from this small aperture rubber latex as described in

Example 1 of U.S. Patent Application Serial No. 09/581,804. The swelling index of the large aperture rubber latex was 8.

B. Polybutadiene 2

Large aperture rubber latex was produced according to the process of Example 1 of U.S. Patent Application Serial No. 09/581,804. The swelling index of the large aperture rubber latex was 21.

C. Polybutadiene 3

Small aperture rubber latex was produced according to the process of Example 1 of U.S. Patent Application Serial No. 09/581,804, except that the reaction was performed at 60°C rather than 65°C and tertiary dodecyl mercapta (TDDM), which is a molecular weight-controlling agent, was added in an amount of 0.4 parts by weight rather than 0.3 parts by weight. Large aperture rubber latex then was produced from this small aperture rubber latex as described in Example 1 of U.S. Patent Application Serial No. 09/581,804. The swelling index of the large aperture rubber latex was 40.

Thermoplastic resins were produced by performing graft copolymerization based on the same materials and processes of Example 1 of U.S. Patent Application Serial No. 09/581,804. Table 2 presents the relationship between the swelling index of the large aperture polybutadiene used to produce the resins and the haze values of the resins produced from the various types of large aperture polybutadiene.

Table 2.

	Swelling Index	Haze Value (%)
Polybutadiene 1	8	2.0
Polybutadiene 2	21	2.0
Polybutadiene 3	40	6.2

When large aperture polybutadiene has a swelling index of 40, which is outside of the range claimed in U.S. Patent Application Serial No. 09/581,804, the haze value of the resin is high. Such a resin is poor in transparency and thus is not suitable for use as a transparent thermoplastic resin.

6. The Examiner has asserted that:

[i]n view of the fact that the refraction coefficient of methyl methacrylate, styrene and acrylonitrile are 1.49, 1.59 and 1.518, respectively, and in view of the fact that the amounts of components decreases in the order, methyl methacrylate, styrene and acrylonitrile, it is highly likely that the refraction coefficient [of the cited prior art composition of Yumoto *et al.*] lies within the claimed range.

The Examiner then states that the burden is shifted to the Applicants to establish an unobvious difference between the cited art and the present invention.

I declare that the Examiner's assessment of the total refraction coefficient of the presently claimed composition, excluding the polybutadiene rubber latex, is incorrect. Amended Claims 1 and 8 of the present invention relate to a thermoplastic resin composition comprising, *inter alia*, (i) 40-70 parts by weight of a methacrylic acid alkylester compound or an acrylic acid alkylester compound, (ii) 15-30 parts by weight of an aromatic vinyl compound, and (iii) 1-20 parts by weight of a vinylcyan compound, wherein a total refraction coefficient of the

composition, excluding polybutadiene rubber latex, is between 1.510 and 1.526. The refraction coefficient of the components above can be calculated using the following formula:

$$1.49 X_A + 1.59 X_B + 1.518 X_C = \text{Refraction Coefficient of Polymer}$$

In the formula above, X_A , X_B and X_C are the weight ratios of methyl methacrylate, styrene, and acrylonitrile, respectively. As indicated by the Examiner, the refraction coefficients of methyl methacrylate, styrene, and acrylonitrile are 1.49, 1.59, and 1.518, respectively. If the weight ratios of methyl methacrylate, styrene, and acrylonitrile are 40/71, 30/71, and 1/71 part by weight, respectively, the refraction coefficient of the polymer of these three components exceeds 1.53, and the presently claimed range. Therefore, the Examiner's assertion that the total refraction coefficient will likely fall within the claimed range is incorrect.

I further declare that the claimed property, *i.e.*, total refraction coefficient of the composition, excluding the polybutadiene rubber latex, between 1.510 and 1.526, is required to achieve a thermoplastic resin composition with the desired transparency. The refraction coefficient of the polymer of the components above has a direct influence on the transparency of the thermoplastic resin composition (*see* page 5, lines 18-19 of the specification of the instant application) and is regulated by the mixture ratio of the monomers. Furthermore, since the refraction coefficient of polybutadiene is approximately 1.518, the refraction coefficient of the remaining grafted components must fall in the claimed range to produce a composition of the desired high level of transparency (*see* page 5, lines 20-22 of the specification of the instant application).

In fact, there is no teaching in Yumoto *et al.*, JP 56-41216, or Kim *et al.*, either alone or in combination one with another, that the total refraction coefficient of the composition,

excluding the polybutadiene rubber latex, must fall within the claimed range of between 1.510 and 1.526. Since the cited art, alone or in combination, fails to teach each and every element of the claimed invention, I declare that Claims 1, 2 and 5-11 are not rendered obvious to the artisan of ordinary skill by the teachings of Yumoto *et al.*, JP 56-41216, and Kim *et al.*

The transparency of the resin improves as the refraction coefficient that is used in the production of the transparent thermoplastic resin becomes smaller. Polybutadiene, which is used as rubber latex in the claimed invention, has a refraction coefficient of approximately 1.518. However, since styrene monomer and butadiene have refraction coefficients of 1.590 and 1.518, respectively, the refraction coefficient of the styrene-butadiene copolymer in Yumoto *et al.* is significantly larger than that of the polybutadiene of the claimed invention. Accordingly, the claimed invention produces a resin that has superior transparency to that of Yumoto *et al.*

The relationship between the refraction coefficient and transparency is directly demonstrated by the following experimental evidence. Three types of thermoplastic resins were produced by graft copolymerization according to the processes of Example 1 of U.S. Patent Application Serial No. 09/581,804 except that the following three types of rubbers were used as the source of the large aperture rubber latex:

- A. Butadiene rubber (refraction coefficient = 1.518)
- B. Styrene-Butadiene rubber (Styrene:Butadiene = 16.2:83.8, refraction coefficient = 1.529)
- C. Styrene-Butadiene rubber (Styrene:Butadiene = 29.7:70.3, refraction coefficient = 1.539)

Table 3 presents the relationship between the refraction coefficients of the large aperture rubbers used to produce the resins and the haze values of the resins produced from the various types of

large aperture rubber.

Table 3.

	Refraction Coefficient	Haze Value
Butadiene	1.518	2.0
Styrene:Butadiene (16.2:83.8)	1.529	3.8
Styrene:Butadiene (29.7:70.3)	1.539	4.9

When large aperture polybutadiene has a refraction coefficient greater than 1.526, which is the upper limit of the range claimed in U.S. Patent Application Serial No. 09/581,804, the haze value of the resin is too high for use as a transparent thermoplastic resin.

7. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of any patent issuing from the above-captioned patent application.

Dated: September 20, 2004

By:

Chan Hong Lee

Dr. Chan-Hong Lee